Peculiarities of Particulate ^{137}Cs Transport and Sedimentation in Kiev Reservoir

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Abstract. The paper presents the data on Chernobyl radiocaesium bound to suspended matter and bottom sediments at different locations along the sampling rout from Rivers of Chernobyl zone to upper Reservoirs of Dnieper River. These data were collected as a result of joint Ukrainian-Italian field exercises in the frame of ECP-3 project. It was found out that total ^{137}Cs concentration in the water column decreases downstream the Chernobyl zone while K_D *in situ* values substantially increase with approach to the Kiev HPS dam. Taking account of uniform hydro-chemical conditions in investigated area one can explain this phenomenon only by gradual elimination of coarse sandy component with low sorption capacity from the river flow by sedimentation. In contrary, radiocaesium which is selectively sorbed and fixed on fine clay particles travels much longer distances and ensures observed higher K_D *in situ* values. This conclusion is supported by the analyses of three sediment cores taken in upper, middle and lower parts of Kiev reservoir.

1. Introduction

An exclusive role of Kiev reservoir as a huge natural trap for Chernobyl radionuclides is defined by its geographical position and hydrological regime. Actually it is an about hundred kilometres long watercourse from the Pripyat river (which is the main carrier of radionuclides from the Chernobyl zone) and the Dnieper river confluence to the HPS dam in the Northern suburbs of Kiev City. According to previous observations in 1986-1993, up to 60% of particulate cesium (i.e. 18-25% of total activity) coming into the Kiev reservoir was trapped and buried in its bottom sediments [1].

In order to better understand the transport mechanisms and to validate models it was extremely important to trace out the particulate ^{137}Cs fate for different size fractions of suspended particles through the pathway from the Chernobyl zone to the Dnieper River reservoirs. This is done by focusing on the key transport parameter K_D distribution coefficient *in situ* which is defined as the ratio between concentrations of radiocesium in particulate [Bq/kg] and soluble [Bq/l] phases. In this particular study it was done it two ways. Firstly, water sampling survey downstream of the Chernobyl zone presents "an instant picture" of particulate cesium transport, and secondly, bottom sediment cores taken at three characteristic point give an idea of temporal variations in suspended cesium concentration in different parts of the Kiev reservoir.

2. Materials and Methods

Samples of water and different fractions of suspended materials were collected in 1992, 1994 and 1995 in two reservoirs (Kanev and Kiev) and four rivers (Dnieper, Pripyat, Uzh and Ilya) which are different in morphological scale and hydrological regime caused in different level of suspended particles contamination. Subordination sequence of the hydrological relationship between above rivers and reservoirs are as follows :

Ilya → Uzh → Pripyat → Dnieper (Kiev res.) → Dnieper → Dnieper (Kanev res.)

Sampling exercise was carried out using two different sampling devices.

The first system was capable to perform both size fractionation of suspended materials and concentration of dissolved caesium directly at time of taking sample. The different fractions of suspended materials were separated on sequentially arranged cartridges with nylon filters (PALL filters, HDC II, 1000, diameter 60 mm, filtration area 0.49 m^2). Filters were used with nominal pore sizes 40 μm, 10 μm and 0.45 μm. The suspended matter mass determined as difference of dried filters weight before and after filtration.

Dissolved ^{137}Cs was concentrated on columns filled in ammonium hexocyanocobaltferrate (NCFN) ion-exchange resins. In order to assure the resin efficiency to concentrate radiocaesium, series of two ion-exchange columns were used (diameter 20 mm, height 160 mm and 80 mm, respectively). The volume of filtered water ranged from 500 to 2000 litres, and samples in each sampling point were taken in two replicates.

The second device was based on a "Millipore" Tangential filtration and allowed to collect the suspended material using cartridge of filters with 0.1 μm pore size. Dissolved radiocaesium was also concentrated on ion-exchange columns described above. Volume of filtered water ranged from 500 to 800 litres.

All filters and resins were subjected for gamma spectrometry using HPGe detectors. The time of sample counting 20 h provided a standard deviation $<10\%$.

An automatically operated logger was continuously used to record changes of pH and turbidity (suspended material content) of water during the sampling time 8 hrs. Some components (e.g. Ca, Mg, Na, K and NH_4) were measured in the laboratory.

One sediment core was taken using 1 meter long mini-Mackereth pneumatic corer in the upper part of Kiev reservoir at the Pripyat mouth (Strakholesye village, 56 km upstream the Kiev HPS dam) and two other ones - by mechanical ADT (30 cm long) corer near Sukholutchye village (37 km upstream the Kiev HPS dam) and in old river channel (18 km upstream the Kiev HPS dam). The location of sampling points is fitted to the old river channel and characterised by very high sedimentation rates (about 4, 3 and 2.3 cm per annum, respectively). The cores were sliced in intervals of 1 or 2 cm, dried and γ-counted in a standard geometry.

3. Results and Discussion

It is noted worthy that hydrochemical conditions along the sampling route turned out to be fairly uniform. Typical values for Ca^{+2} were reported as 50 mg/l, for Mg^{+2} - 10 mg/l, for Na^{+} - 15 mg/l and for K^{+} - 3 mg/l; pH values ranged from 8 to 8.5 except for the Ilya river (sampling point 11) where it was registered as low as 6.9. Nevertheless the difference in hydrological conditions in rivers and reservoirs stipulated drastic variations in the results related to suspended radiocaesium.

Table 1 shows the ^{137}Cs concentrations in water and ^{137}Cs distribution coefficient (K_D) calculated "*in situ*" at each sampling site for the different grain sizes of suspended material. At all sampling sites ^{137}Cs seems to be more strongly associated with the largest size fractions of suspended matter (40 and 10 μm). These results appear surprising and could be attributable to the high bloom of algae observed during the sampling periods (June-July). *To explain the higher values of* ^{137}Cs *found in the largest size fractions of* suspended matter (40 and 10 μm) the following hypothesis has been formulated : algae surfaces could present natural traps for the finest suspended particles. In this way the material collected in the filters of 40 and 10 μm pore sizes could be affected by the presence of mineral fractions and this could justify the higher values of caesium K_D found. To clarify the role of phytoplankton the uptake of radionuclides and role of suspended material adhesion onto surfaces of algae and aquatic plants, a more detailed experiment has been planned.

Table 1. ^{137}Cs Dissolved in Water and K_D *in situ* Values (1993) in the Different Fractions of Suspended Material

Sampling Point	#	Susp. Part. (mg/l)	^{137}Cs *in* water (mBq/l)	"In situ" K_D $*10^{-3}$ (l/kg)			
				Total	40 μm	10 μm	0.45 μm
Illya River (2.5 km upstr. Uzh)	11	7	420±10	8±1	23±1	9±1	3.2±0.2
Uzh River (15 km upstr. Pripyat)	10	10	73±2	50±6	70±8	38±6	20±1
Pripyat River (Chernobyl)	9	22	70±10	69±33	74±36	44±13	17±5
Dnieper River (10 km upstr.Kiev res.)	7	18	16±1	108±2	106±5	181±59	22±5
Kiev Reservoir (Pripyat&Dnieper confl.)	6	8	90±10	79±4	90±8	73±16	28±1
Kiev Reservoir (45 km upstr. Kiev dam)	8	4	106	106	124	100	47
Kiev Reservoir (25 km upstr. Kiev dam)	4	3	94±2	113±1	143±7	103±1	50±6
Kiev Reservoir (3.5 km upstr. Kiev dam)	2	3	74±1	87±3	109±1	69±15	64±14
Dnieper River (Kiev City)	3	9	50±3	69±2	77±4	64±3	39±1
Kanev Reservoir (68 km dnstr. Kiev dam)	5	6	45±1	79±5	91±5	79±4	53±3

Since the 40 and 10 μm filters may have been affected by the presence of mineral fractions, only the data relating to the fraction retained on the 0.45 μm filters are discussed.

It can be clearly seen from the data that when downstream the Pripyat river and Kiev reservoir the water velocity in the main channel slows down, suspended matter content decreases and radiocesium K_D *in situ* increases to the highest values in the lower part of the reservoir (p.2). Further down the Kiev HPS dam in the Dnieper river channel (p.3) the K_D value substantially decreases and then increases again in the lower part of Kanev reservoir (p.5).

The most appropriate explanation of this phenomenon seems to be a drastic change in the mineral composition within the finest fraction along the sampling route. It appears that heavier sandy inclusions with low sorption capacity are mostly deposited near the Pripyat river mouth in upper part of Kiev reservoir and further according to formation of sedimentation "cone". One the contrary finer clay fractions travel down the main channel and ensure higher K_D values in lower parts of the reservoir.

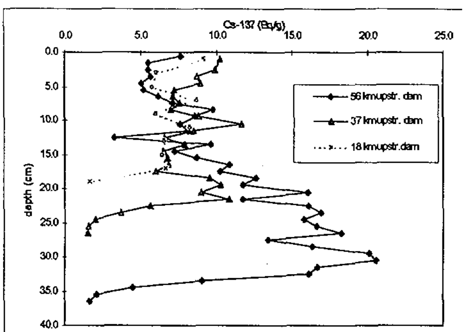

Fig.1. ^{137}C profiles in bottom sediment at different parts of Kiev Reservoir

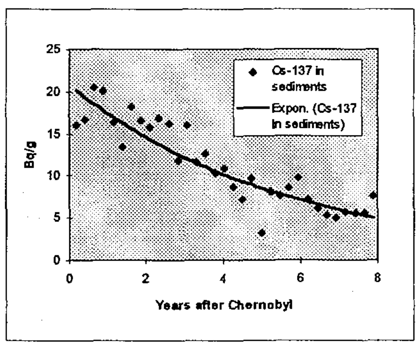

Fig.2. Exponential time trend for dated core from upper part of Kiev Reservoir

This hypothesis is also confirmed by the results of measurements of bottom sediment cores presented in Fig. 1. It shows much higher sedimentation rate near the Pripyat river mouth and substantial decrease of radiocesium concentration in sediment in recent years (probably because of river channel flushing). When plotted versus time of deposition (Fig.2) "post-Chernobyl" part of this profile shows the trend $exp(0.18t)$ with $R^2=0.76$, where t is time measured in years after the accident. On the other hand similar regression for total ^{137}Cs concentration in water in the Pripyat river near Chernobyl for the same period (2500 records corrected to 1986) gives $exp(-0.21t)$ with $R^2=0.81$ [2]. So, it seems that in the upper part of the reservoir near the Pripyat river mouth, the concentration of particulate radiocesium is determined by the concentration in river water. On the contrary looking at the profiles from the middle and upper parts of the reservoir one can conclude that here particulate concentration is more or less stable and defined by an irreversible contamination of fine clay material eroded from the Pripyat catchment [3].

In conclusion, a very important practical implication from the analysis of the presented data could be formulated. Modelling of radiocesium transport in the river-reservoir system with suspended matter should be based on detailed knowledge about changes in mineral content of each size fraction and mechanisms of their contamination (i.e. selective and non-selective sorption, fixation, etc.) and consequent K_d variation along the water course. In this regard a standard approach which implies use of constant K_d values for some different size fractions [4] seems to be very simplified and somewhat misleading.

References

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